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Thermoluminescence of NaCl and NaCl : Rb

S Dorendrajit Singh[1]*, W Shambhunath Singh[2] and P S Mazumdar[3]

[1]Physics Department, Manipur University, Canchipur-795 003, Manipur, India

[2]Physics Department, Manipur College, Singjamei-795 008, Imphal, Manipur, India

[3]Acharya Prafulla Chandra College, New Barrackpur-743 276, West Bengal, India

E-mail : S_dorendrajit@rediffmail.com

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Abstract : Comparison of optical absorption spectrum and TL of pure NaCl and NaCl : Rb (0.7% by weight) is made. The composite TL peaks of pure and doped NaCl are deconvoluted using both general order (GO) and mixed order (MO) kinetics model giving the trap parameters. The effect of optical bleaching of γ-irradiated NaCl is discussed. The growth curve of pure NaCl is presented.

Keywords : Thermoluminescence, general order kinetics, mixed order kinetics.

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General order (GO) [1] or mixed order (MO) kinetics [2] are used to analyse thermoluminescence (TL) peaks. MO kinetics model has been developed by Chen *et al* [2] from the set of three differential equations [3]. May and Patridge [4] were the first to propose the empirical GO equation for the process as

$$I(t) = -\frac{dn}{dt} = n^b s' \exp\ (\text{-}E/kT) \tag{1}$$

where n is the concentration of trapped electrons at any instant of time t, b is the order of kinetics, s' is the pre-exponential factor, E is the activation energy, k is the Boltzmann constant and T is the absolute temperature, Gartia *et al* [5] introduced the filling factor $f = n_0/N$ (where n_0 is the initial concentration of trapped electrons and N is the total number of traps) in the form

$$I(t) = Nf^b s \exp\ (\text{-}E/kT) \tag{2}$$

which can subsequently be used as a mathematical tool in dosimetry since f is directly related to dose. A TL intensity equation without the empirical parameter b has been obtained by Chen *et al* [2] using the MO kinetics model as

$$I(t) = -dn/dt = s_1\, n(n+c) \exp\ (\text{-}E/kT) \tag{3}$$

$$\text{with } s_1' = sA_m / NA_m \tag{4}$$

where A_m, A_n are the recombination and retrapping probabilities and c is the concentration of trapped electrons or holes not taking part in the TL process in the temperature range considered due to their being in deep traps or in low probability recombination centers.

Solving eq. (3) under the linear heating profile $T = T_0 + \beta t$, (β being the heating rate), one gets

$$n = n_0(1-\alpha) \Big/ \left[\exp\left\{ (cs_1/\beta) \int_{T_0}^{T} \exp\ (-E/kT')dT' \right\} - \alpha \right] \tag{5}$$

where $\alpha = n_0/(n_0 + c)$ and T_0 is the temperature at time $t = 0$. Differentiating eq. (5) with respect to T, the expression for TL intensity becomes

*Corresponding Author

$$I(T) = \frac{Nf^2(1-\alpha)s''\exp(-u)\exp\left[(fs''E/k\beta)\{E_2(u)/u - E_2(u_0)/u_0\}\right]}{\left[\exp\{(fs''E/k\beta)(E_2(u)/u - E_2(u_0)/u_0)\} - \alpha\right]^2} \quad (6)$$

where $s'' = (1-\alpha)sA_m/\alpha A_n$. (7)

Here, $u = E/kT$, $u_0 = E/kT_0$ and the temperature integrals have been expressed in terms of second exponential integral [5].

Yossian and Horowitz [6] annealed the irradiated LiF : Mg, Ti (TLD–100) at 165°C to remove the low temperature peaks. The isolotaed TL peak V has been fitted using MO kinetics model. Singh *et al* [7,8] have presented a new set of expressions for the determination of activation energy E using mixed order parameter α. TL glow curves of γ-irradiated NaCl : I (2.04 KGy) are recorded (linear heating rate 1.5°C s^{-1}) after thermal clearing at 169°C and 182°C. The curves have been fitted with the MO model. Similarly, TL of 5 minutes X-ray irradiated BeO (maximum temperature, T_m = 160.1°C) has been fitted with the MO formalism [9].

The present paper attempts to analyse TL peaks of pure NaCl and NaCl : Rb (0.07 by weight). Optical absorption spectrum of identical samples of pure and NaCl : Rb are presented. In this paper, curve fitting technique [10] using MO and GO model is used to find the activation energies E, frequency factor, the MO parameter α of TL peaks of pure NaCl and NaCl : Rb. A good correlation has been obtained between the order of kinetics b and the MO parameter α.

The samples of approximate size 4 x 4 x 1 mm^3 are cleaved from the same bulk of crystals of pure NaCl and Rb (0.7% by weight) doped NaCl. The samples are irradiated by γ-rays of different doses ranging from 0.080–3.030 kGy.

Comparison of optical absorption spectrum of identical samples of pure NaCl and NaCl : Rb both irradiated by γ-ray (1.01 kGy) in the wavelength range of 200-800 nm is shown in Figure 1. Both curves show a broad band around a peak occurring at wavelengths 467 nm and 461 nm in respect of pure NaCl and NaCl : Rb. The broad band at around 460 nm for both samples are due to the presence of F centres and its other possible forms [11]. However, since all possible forms of F centres have different thermal stabilities, and the experiment is carried out at room temperature, the absorption spectrum shows a broad band disguising the presence of separate forms of F centres.

The deconvolution of composite TL peaks of pure NaCl (Figure 2) and NaCl : Rb (Figure 3) irradiated by γ-rays (1.01 kGy) by using MO kinetics model shows that TL of both samples comprises three component peaks

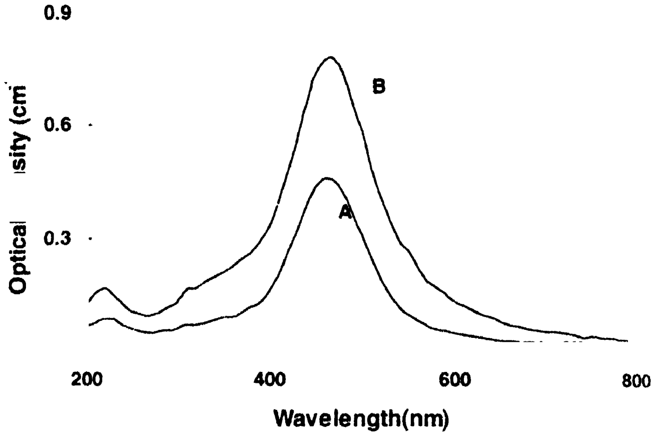

Fig 1. Optical absorption spectrum of pure NaCl (marked A) and NaCl : Rb (marked B) irradiated by γ-rays (1.01 kGy).

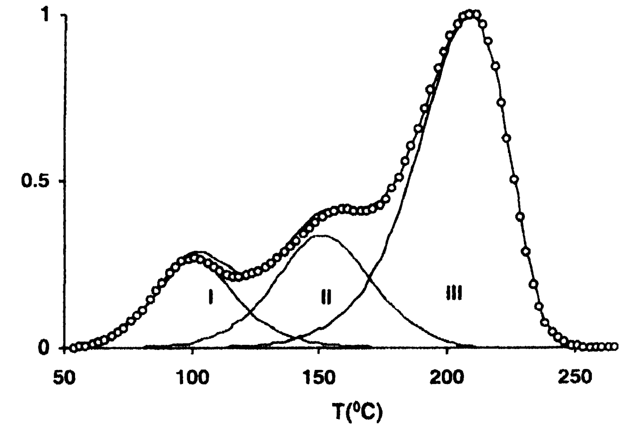

Fig 2. Curve fitting of TL peak of pure NaCl irradiated with γ-rays (1.01 kGy) recorded without fading using MO kinetics model. The peaks I, II and III correspond to activation energies 1.09 eV, 1.12 eV and 1.16 eV; values of α as 1.0, 0.89 and 0.01 and s'' as 10181 s^{-1}, 654 s^{-1} and 1.7 s^{-1} respectively. The value of N has been taken to be equal to 10^{10} cm^{-3} and F = 1 for all the three peaks considered.

The experimental peaks of NaCl and NaCl : Rb have also been fitted using GO kinetics model; and three component peaks identical with those obtained using MO kinetics model, are obtained for both. The various MO and GO parameters associated with TL peaks of NaCl and NaCl : Rb are presented in Table 1. In agreement with the observation made by Murti *et al* [12] that a similar electron trap with different perturbations due to environmental differences can give rise to different glow peaks, the TL of pure NaCl has three peaks having approximately the activation energy of 1.1 eV. The observation is slightly different for the case of NaCl : Rb for which the last peak (peak III) is found to be having larger activation energy than those of the rest two peaks. The first peak (peak I) is found to have the same E as that of the pure sample but that of the second peak (peak II) has the least E of all the peaks. Thus, it can be stated that the introduction of impurity ion Rb^+ in the lattice of NaCl has perturbed the F-centres and added two levels of different activation energies.

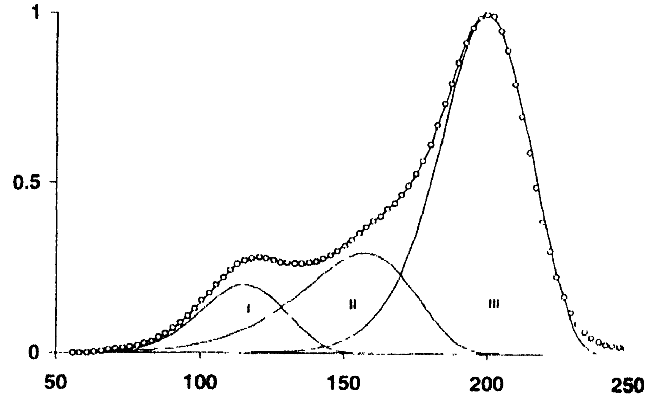

Fig 3. Curve fitting of TL peak of pure NaCl : Rb irradiated with γ-rays (1 01 kGy) recorded without fading using MO kinetics model The peaks I, II and III correspond to activation energies 1 20 eV, 0 78 eV and 1 56 eV; values of α as 0 74, 0.01 and 0 68 and as $10^2\ s^{-1}$, $10^{-5}\ s^{-1}$ and $10^4\ s^{-1}$ respectively The value of N has been taken to be equal to $10^{10}\ cm^{-3}$ and $f = 1$ for all the three peaks considered

The lifetimes τ of the three TL peaks of pure NaCl and NaCl : Rb at ambient temperature $T = 300K$, are estimated using the relation [5]

$$\tau = \frac{1}{s}\exp(E/kT)$$

and are presented in Table 1. The first two peaks of both samples have short life times ranging from few minutes to few days. The last peak (peak III) of both samples are stable having lifetimes in years. Keeping in mind of the longer life time of peak III of NaCl of 21 years (Table 1) compared to those of peak I and II, the growth of peak III is studied. Samples of NaCl cleaved with approximate size of 4 x 4 x 1 mm^3 but having identical masses are irradiated with γ-rays having doses of 0.080, 0 168, 0 505, 1.010, 1.515 and 3.030 kGy and their read-outs are recorded with the same heating rate of 1.95°C s^{-1} after 6 days fading. The maximum intensity I_m of the peak III has been plotted with various doses of γ-rays. TL growth of peak III of pure

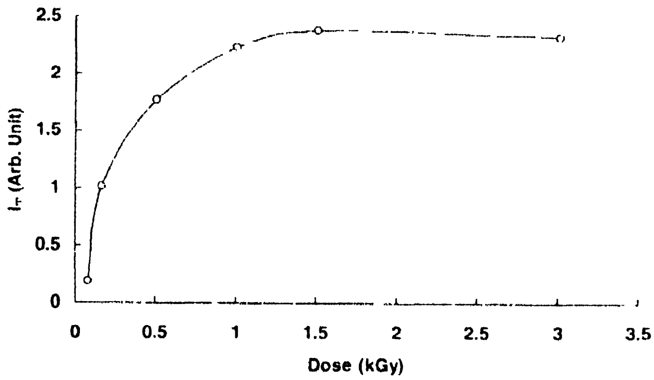

Fig. 4. TL growth of the peak III of pure NaCl Open circles represent the observed peak intensity I_m of peak III of pure NaCl

NaCl is presented in Figure 4. It is observed that the growth is quadratic (with square of correlation coefficient ~ 0.95) upto a dose of around 1.515 kGy after which saturation occurs. The present observation agrees well with the model of TL growth formulated by Kristianpoller *et al* [13], which predicts a quadratic super linear TL growth with dose. The growth curve of peak III behaves in a similar manner with the growth of F centres of pure NaCl [14]

Table 1. MO and GO parameters associated with component peaks of NaCl and NaCl Rb irradiated by γ-rays (1 01 kGy)

Model	Prameters	Pure NaCl			NaCl Rb		
		Peak I	Peak II	Peak III	Peak I	Peak II	Peak III
Mixed order	T_m(°C)	100 62	151.00	208 58	115.43	157 66	199 70
kinetics	E(eV)	1 09	1.12	1.16	1 2	0 78	1 56
$N = 10^{10} cm^{-3}$	α	1.00	0 89	0.01	0 74	0.01	0 68
$f = 1$	s'' (s^{-1})	10^4	1.0	2.0	10^2	10^{-5}	10^4
	T_m(°C)	99 5	151.04	208 61	115.52	157 67	199.89
General order	E(eV)	1.19	1.11	1.12	1 1	0.79	1.45
kinetics	b	2.33	1 72	1 00	1.46	1.00	1.38
	s' (s^{-1})	4.1×10^{17}	9.9×10^{13}	$9\ 3\times10^{9}$	3.5×10^{13}	1.7×10^{8}	$3\ 9\times10^{14}$
	τ	16mts.	12hrs.	21yrs	4days	2days	4000yrs.

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